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Fischer, P

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Magnetic imaging with polarized soft X-rays

Peter Fischer

Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley CA 94720, USA
and
Physics Department, UC Santa Cruz, Santa Cruz CA 95064, USA
PJFischer@lbl.gov

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Abstract

Properties, behavior, and functionalities of magnetic materials are largely determined by microscopic spin textures, particularly their formation into domains, their coupling mechanisms and their dynamic behavior. Advanced characterization tools are prerequisite to fundamentally understand magnetic materials and control spins for novel magnetic applications. Magnetic microscopies allow to image directly the static and dynamic features of the relevant microscopic magnetization structures in advanced magnetic materials and thus provide detailed and direct insight into underlying physical phenomena. A large variety of magnetic imaging techniques has become available with particular strengths but also certain limitations.

Essential features of magnetic microscopies are a high spatial resolution down into the nanometer regime, as this is the fundamental length scale of magnetic exchange interaction and the ultimate length scale in advanced magnetic technologies; magnetic and elemental sensitivity with quantitative capabilities, as the properties of advanced magnetic materials can be tailored by combining various magnetic elements and their magnetic moments; high temporal resolution from the nsec to the fsec regime to understand the associated spin dynamic processes and the functionality in magnetic devices; tomographic capabilities with nm resolution as new directions in nanoscience and technologies are moving into 3dim arrangements of spin structures; and interfacial sensitivity as novel ways to control spins harness either the coupling across interfaces in multilayered structures or utilize non-collinear spin arrangements, which often occur from symmetry breaking at surfaces and interfaces.

The unique properties of polarized soft x-rays, their abundancy and specific interaction with magnetic materials in form of dichroism effects have triggered the development of various magnetic x-ray imaging techniques. This review will provide an overview of the current state-of-the-art in magnetic imaging with polarized soft x-rays. It describes the various approaches using x-ray optics, electron optics and diffraction based techniques and it will highlight the capabilities of each technique by selected examples from current research.

1. Introduction

Since ancient times magnetic materials have played an important role in our life. Despite the fact, that for centuries the phenomenon of magnetism carried some kind of mystical character, the use of magnets allowed to navigate and explore the globe, and permanent magnets in electric motors and generators were key components in the industrial revolution. Today, magnets are ubiquitously and found in a wide range applications from generation, storage, and transmission of energy in the large scale power grid to the nanoscale world of information and sensor technologies, and from future environmentally conscious mass transportation systems to magnetic resonance imaging (MRI) diagnosis in the health sector.

Scientifically, the magnetism in materials continues to be one of the most challenging and rewarding topics in materials sciences [1]. The fundamental unit in magnetism is the spin of the electron. Similar to electronics, where the charge of the electron is the ultimate quantity determining behavior and functionality of devices, spintronics devices relies on the spin of the electron [2]. Understanding and controlling the static properties, dynamic behavior and the functionalities of magnetic materials is fundamentally connected to spin textures that range from the nanoscale to the mesoscale [3], [4] . Magnetic interactions, such as magnetic exchange and anisotropy define the associated energies and the resulting spin textures display the ground and excited states.

Imaging magnetic materials is one of the most valuable characterization tool and therefore a tremendous effort is being put into developing appropriate imaging techniques. The impact of magnetic microscopies to a fundamental understanding of magnetic materials can be measured by achievements in the following categories.

- **Spatial resolution:** Fundamental length scales in magnetism are determined by exchange lengths. They determine e.g. the scale and the distance over which spins can change their orientation, such as in domain walls or vortex cores. Although material dependent, typically exchange lengths are in the few nanometer regime, which is therefore the targeted spatial resolution of advanced magnetic microscopies. Topology in spin textures plays a major role for novel and unconventional spin structures, and even more non-trivial topologies that can emerge, e.g. in multiple spatial dimensions (3D tomography).
- **Temporal resolution:** Fundamental magnetic time scales are related to the strongest magnetic interactions, i.e. again the exchange energy, from which the fsec regime derives. However, it is the dynamics across multiple time scales from fsec to years that will determine the behavior and enables the control of magnetic materials.
- **Elemental, chemical, and interfacial sensitivity:** In the search for magnetic materials that will exhibit novel properties, behavior and functionalities, multicomponent materials serve as the primary design principle. It is the interfacing of different materials thereby harnessing proximity and confinement principles that enable new magnetic features. Starting points can be either a theoretical prediction or a computational design of novel magnetic materials, or the actual synthesis of complex material. Characterization, in particular imaging tools that allow to distinguish the

impact of each individual component and that have direct access to buried interfaces are critical in verifying both the theoretical and computational models and the advanced synthesis.

- **Quantifiable magnetic information**: Beyond purely resolving spin structures, it is specifically for applications of magnetic materials important to be able to quantify magnetic properties. A direct measurement of those quantities is very desirable, notably if they can be assigned to individual components.
- **Imaging in various environments:** Imaging magnetic materials and structures as a function of external parameters, including magnetic and electric fields, temperature, pressure will provide detailed insight into their dynamics and behavior.

Given the importance of imaging magnetic structures and systems, numerous approaches to advance magnetic imaging techniques are being investigated and developed. They can be broadly categorized into the various probes they are using. In comparison with the polarized soft x-rays, which are the sole focus topic of this review, all of these techniques have their pros and cons. For example, atomic spatial resolution has been documented with spin-polarized Scanning Tunneling Microscopy (SP-STM), but it is difficult to obtain ultrafast time resolution with SP-STM. Likewise, magneto-optical microscopies using ultrafast optical laser systems regularly demonstrate unprecedented time resolution, but due to the wavelength of optical light, their spatial resolution is limited. The following list is a very brief description of those broad categories of magnetic imaging techniques including some key literature that should help the reader to become familiar with the main concepts.

- **Magneto-Optical microscopies**: Those techniques take advantage of the various magneto-optical effects, where the polarization state of light is slightly altered by the magnetization state of the specimen. The effects include the Kerr-effect, the Faraday-effect, the Voigt effect, and the Cotton-Mouton effect [5]. The currently available optical laser systems make those MO microscopies a very versatile tool with an excellent time resolution (fsec), however, the limited penetration depth and the size of optical wavelengths limits its spatial resolution (typically a few 100nm) severely from reaching into the nanoscale regime and into complex 3D magnetic systems, resp.. Furthermore, elemental and chemical sensitivities are limited due to the rather unspecified nature of optical transitions in the valence bands.
- **Magnetic scanning probe microscopies**: This category spans probably the largest range [6]. It encompasses Spin polarized Scanning Tunneling Microscopy[7], [8], which allows not only to image spin textures at nearly atomic resolution [9], but has recently been able to expand in the time domain so as to study e.g. fast electron spin relaxation times occurring in the ns regime [10]. There is also the very diverse field of scanning force microscopies, where the highly versatile magnetic force microscopes (MFM) are abundantly used for magnetic imaging down to better than 10nm spatial resolution. A most recent development with high potential is Nitrogen Vacancy (NV) center scanning probe microscopy [11] that provides quantitative and high sensitive measurements of the stray magnetic field emanating from a nanoscale spin texture. Ultimately, the spatial resolution with NV center microscopies is limited by the atomic size of the probe. A recent achievement was the successful detection of the stray field from a single electron spin [12].
- **Magnetic electron microscopies**: They utilize the interaction of electrons with the magnetization of the

sample. This can be either spin dependent scattering or the Lorentz force acting on the electrons. The former interaction is e.g. the basic principle in scanning electron microscopy with polarization analysis (SEMPA) [13] or spin polarized low energy electron microscopy (SPLEEM) [14]. Both those techniques are highly surface sensitive. The latter interaction, i.e. the Lorentz force is used in Lorentz transmission electron microscopes (L-TEM) [15], where the limited penetration of electrons requires the specimen to be relatively thin. However, this matches well with scientifically interesting and technologically relevant thin film magnetic systems. Recent developments include magnetic 3D tomography with LTEM [16], and highest spatial resolution can be obtained with aberration corrected electron microscopes (sub-nm regime). A very interesting direction for electron microscopy are tailored modifications of the electron wave front, so called vortex beams that carry significant orbital angular momentum [17]. The use of AOM in novel studies of magnetic materials is still in a nascent state. Electron holography is used to image the magnetic texture by recording an interference pattern between the original beam and a reference beam [18]. Finally, there are significant recent efforts to push electron diffraction, including electron microscopy into the ultrafast (psec to fsec) time regime [19].

- **Magnetic x-ray microscopies**: The properties of x-rays give those techniques a unique combination of features that are highly relevant to magnetic imaging [20]. This topical review provides an overview of x-ray microscopies that are currently being used or developed for magnetic imaging. First, it will briefly describe the magnetic contrast mechanisms, which are commonly used by the various x-ray imaging techniques, followed by a short description of the experimental techniques. Selected examples from recent research will demonstrate the current state-of-the-art accomplishments to meet the above mentioned features and detail some of the scientific highlights. Finally, an outlook is given into future opportunities and challenges with magnetic x-ray microscopies.

2. Principles of magnetic contrast with x-rays

The physical phenomenon underlying magnetic imaging technique using x-rays are various dichroism effects [21]. For the study of ferro- and ferromagnetic materials it is primarily X-ray Magnetic Circular Dichroism (XMCD) [22] that is sensitive to the magnetization $\langle \mathbf{M} \rangle$, whereas for antiferromagnetic materials the analogous X-ray Magnetic Linear Dichroism (XMLD) [23] effect that is sensitive to $\langle \mathbf{M}^2 \rangle$ are being used as magnetic contrast mechanisms.

XMCD can be seen as the x-ray counterpart of the magneto-optical Kerr and Faraday effects. Circular polarized light interacts with the magnetization of a ferromagnetic species and leads to a helicity dependent absorption of the x-rays depending on the relative orientation of the x-ray helicity to the projection of the magnetization onto the photon beam propagation direction. The XMCD effect can be understood by the following model (see Fig. 1). A circularly polarized photon with an energy matching the binding energy of an inner core atomic level generates a spin and orbital polarized photoelectron inside the solid that is transferred into an unoccupied spin and orbital dependent state at the Fermi level. In case of the spin states, there are majority and minority states with different density of states at the Fermi level, and therefore the absorption coefficient describing the probability to undergo a certain dipolar transition from the unpolarized inner-core level to the unoccupied state at the Fermi level is different, i.e. there is a magnetic dichroism effect that can be used as magnetic contrast. Angular momentum conservation and spin-orbit coupling in the initial state lead to a reversed XMCD effect at

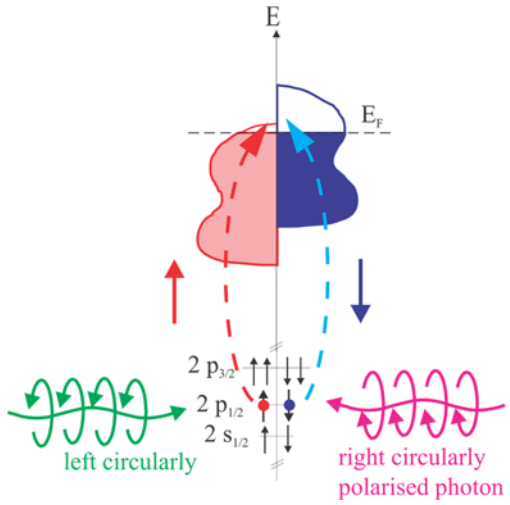

Figure 1: Schematic description of the absorption process of a circularly polarized x-ray at the L_2 absorption edge.

spin-orbit coupled core states, such as the $2p_{1/2}$ and $2p_{3/2}$ levels, which correspond to the $L_{2,3}$ absorption edges. The photon energies for those $L_{2,3}$ edges in 3d transition metals, such as Fe, Co, Ni are in the soft x-ray regime. Note, that the corresponding K-edges for Fe, Co, Ni, i.e. the transitions from $s_{1/2}$ levels to p levels are in the hard x-ray regime. Although the first XMCD studies were performed at those K-edges of Fe, the XMCD effects are much smaller as the s-electron in the initial state has no orbital moment and therefore the XMCD originates from a weak coupling in the final state. Further, since the much higher penetration of hard x-rays leads to a significantly reduced signal/noise ratio at those hard x-ray K-edges, soft x-ray XMCD studies are strongly favored for magnetic x-ray spectromicroscopy.

For the studies of antiferromagnetic materials, XMLD, is generally be used. This effect also occurs in the vicinity of x-ray absorption edges, but depends on the orientation of the magnetic domains relative to the orientation of the linear polarization of the photons and therefore scales with the absolute value of magnetization only.

Since the x-ray dichroism effects occur predominantly in the vicinity of x-ray absorption edges, the associated inherent elemental specificity is one of the major features of magnetic x-ray spectromicroscopy as it allows to distinguish the magnetic properties and behavior of individual constituents in a multi-component material. The sensitivity of XMCD spectromicroscopy measurements can be enhanced and the magnetic contribution to the spectroscopic signal can be separated form non-magnetic background signals by harnessing either the contrast reversal at spin-orbit coupled edges (see above) or through a modulation of circular polarization [24].

Quantitative information can be derived from the XMCD signal by applying so-called magneto-optical sum rules [25], [26]. Element-specific spin and orbital magnetic moments can thus be obtained in a unique way [27]. Since x-ray spectroscopy is a fingerprint of the underlying band structure of the solid, magnetic moment or at least ratios between spin and orbital contribution can be detected with very high precision.

In addition to the magnetic x-ray absorption contrast, there is also a corresponding magnetic x-ray phase contrast [28]. Although those two effects are linked via a Kramers-Kronig relationship, so far, mostly the absorption part has been exploited by magnetic x-ray spectromicroscopy studies. However, the appearance of future x-ray light sources with a high degree of coherence and increased intensities as well as novel techniques allowing to retrieve the magnetic x-ray phase [29] [30], [31] either experimentally or through calculations with high precision, should foster more phase sensitive magnetic x-ray dichroism studies.

The wavelength of soft x-rays is in the nm regime and for hard x-rays even in the sub-nm range. Compared to optical microscopies, x-ray based imaging will therefore ultimately provide a diffraction limited spatial resolution at the very fundamental magnetic length scales. Below, we will highlight a few examples showing the current state-of-the-art in spatial resolution with magnetic x-ray imaging. Whereas several competing magnetic imaging techniques either have ultimate spatial **OR** temporal resolution, x-ray based spectro-microscopies bear the potential to combine both, i.e. have spatial resolution in the nm range **AND** a time resolution down to the fsec regime. The majority of current time resolved x-ray microscopy studies relies on a pump-probe configuration, since the intensity per x-ray pulse at synchrotron storage rings is insufficient to generate a single image. This situation is different at X-ray Free Electron Laser (XFEL) facilities, and therefore single shot (magnetic) imaging capabilities have emerged there [32]. It is worth noting that apart from time-resolved direct imaging of spin dynamics, studies in the frequency space, e.g. X-ray photo correlation spectroscopy harnessing again the increased coherence at next generation light sources will provide a complementary access into the dynamics of spin systems [33] [34].

Access to magnetic interfaces is possible with x-rays in various ways and there are numerous current instrument developments to achieve this. One approach is to harness x-ray reflectivity, which has demonstrated depth resolution in layered structures [35] [36]. In combination with a lateral resolution from an x-ray microscope, this would provide detailed information about the spin textures and their dynamics at buried interfaces. Another approach is to utilize x-ray standing waves that can be moved vertically through an interface with high precision [37] [38]. Finally, a true x-ray tomography with nm spatial resolution will allow to study the internal spin configuration of a solid, in particular the spin properties and behavior at those interesting interfaces. The examples shown below will present the current status of instrument development.

3. Magnetic x-ray imaging techniques

Generally, there are two classes of x-ray imaging techniques that are either currently being developed or already used in magnetism research. Images of magnetic spin textures can be formed either directly in real space or derived from reciprocal space data.

3.1. X-ray microscopies using x-ray optics

Although x-rays have been discovered more than 100 years ago and their short wavelength was immediately recognized as a potential push towards higher spatial resolution compared to optical microscopies, it was the lack of appropriate x-ray optics that prevented x-ray microscopies for nearly a century from being established. It was not until the mid-1980s when not only x-ray sources from synchrotron storage rings became available, but also the capability of nanopatterning materials by e-beam lithography that was originally developed for semiconductor industry enabled then the fabrication of Fresnel zone plates (FZP), which are now commonly used for focusing x-rays, notably in x-ray microscopes, but also for other future advanced characterization tools, such as nano-Angle Resolved PhotoEmission Spectroscopy (n-ARPES) [40]. FZPs are circular diffraction gratings with a wavelength dependent focus length. The spatial resolution that can be obtained with FZPs is largely determined by the outermost zone width of the FZP, which has been demonstrated to reach into the 10nm regime. The

efficiency of FZPs is mostly related to the aspect ratio and the quality and precision of the individual zones, which poses significant challenges to the fabrication of high spatially resolving FZPs as it is the outermost zones, i.e. the thinnest ones that enable highest spatial resolution but also demand for the largest aspect ratio. . There are several other approaches with X-ray optics aiming to achieve highest spatial resolution as well as high efficiency, such as multilayer Laue lenses [41], refractive lenses [42], or zone-doubled diffractive optics [43], to name but a few.

There are two variants of FZP based x-ray microscopes that are used for magnetic imaging, namely a magnetic full-field transmission soft x-ray microscope (MTXM) [44] and a scanning transmission soft x-ray microscope (STXM) [45]. The optical concept for the TXM is analogous to optical microscopes. At the TXM at the Advanced Light Source in Berkeley (BL 6.1.2), x-rays from a bending magnet are overfilling a condenser zone plate (CZP) that provides a hollow cone illumination of the specimen. Together with its wavelength dependent focal length the CZP combines the monochromatizing and the focusing task into one single element. The photons with a spectral resolution of about 1eV at 1keV energy transmit the sample up to a thickness of about 100-200nm corresponding to the probing depth of TXM, and are then imaged through a microzone plate (MZP), i.e. a high resolution x-ray optics onto a 2 dimensional x-ray detector such as a charge-coupled-device (CCD). So far, spatial resolution as low as 10 nm has been published with state-of-the-art FZPs [46], [47], [48].

To allow for magnetic imaging, circular polarization is selected by blocking the upper or lower half of the x-ray cone from the source by an aperture located upstream the CZP. Depending on the contrast the exposure time for each image is a few seconds covering a typical field-of-view of a few micrometer. Selecting a distinct photon energy, or scanning the photon energy with TXM to do spectromicroscopy, one has to only move the CZP and the MZP along the x-ray optical axis to match their respective wavelength dependent focal lengths.

Synchrotron sources have an inherent time structure, which is determined by the filling patterns of the storage rings with the electron bunches generating the x-rays in the various worldwide synchrotron facilities. In general this time structure is not resolved in static x-ray imaging, but it can be utilized particularly for imaging magnetization dynamics. For time-resolved x-ray microscopy studies pump-probe geometries are employed synchronizing the clock frequency of the storage ring, i.e. the probing x-ray pulses to the excitation pump pulse of the sample and then record images at variable delay time between the electronic pump and the x-ray probe. At the ALS, where most of the presented time-resolved magnetic x-ray imaging studies featured in this article were obtained this approach is mostly performed in the so-called 2-bunch mode of the storage ring, although gated CCDs have demonstrated the ability to select individual bunches from the regular multibunch operation of synchrotrons [49]. At the ALS, the x-ray pulse length that sets the temporal resolution is about 70 ps. The spacing between bunches in multibunch mode is about 2ns, whereas in two-bunch operation mode the two bunches are separated by 328 ns allowing for an easy reset of the spin dynamics into the ground state between consecutive x-ray pulses.

Although most TXMs are located at bending magnet sources, the TXM at BESSY in Berlin/Germany operates at an undulator source [50]. The higher intensity allows to decouple the combined focusing

and the monochromatizing function of the CZP into a high spectrally resolving x-ray monochromator and a separate illumination element such as a glass capillary. A significant increase in spectral resolution ($E/\Delta E$) up to 10,000 has been reported so far, which allowed to distinguish the t_{2g} and e_g symmetry split sublevels in the Ti 3d band of TiO_2 [51].

TXMs have been used extensively for x-ray nanotomography, notably with cell biology research, where the highly transparent water window allows for imaging cells in their natural environment [52]. There, the samples are mounted inside a glass capillary, which is rotated at an axis perpendicular to the beam direction to record a full angular set of projection images of the sample. Computer reconstruction algorithms allow to retrieve the whole cell structure in 3D. A similar approach can be followed to do magnetic x-ray tomography [53], however, the main difference is that for spin systems one needs to retrieve a vector quantity, the spin, from the projections, whereas for the cell tomography one retrieves a scalar quantity, the charge.

The STXM approach uses the high resolution MZP from the TXM as a focusing optics upstream the sample and then raster scans either the focused beam across the sample or scans the sample across the focused beam. Both bending magnets and undulator sources are employed for STXMs, although a polarized undulator beam is desirable in particular for magnetic STXM studies as it has generally a higher degree of circular polarization compared to a bending magnet and the switching of polarization is more accurate than for a bending magnet beamline. The photons transmitted through the sample in a STXM are generally detected by an x-ray point detector, such as an avalanche photo diode. The fast response time of an avalanche photo diode (APD) allows for recording individual bunches from the storage ring and also for single photon counting. Other detection modes such as fluorescence or sample current measurements have been published [54]. Since STXM is essentially a micro-spectroscopy technique, those microscopes are mostly used for spectroscopic mapping. For time resolved magnetic studies, the point detector and the possibility to lock-in the synchronization to the storage ring has recently led to a significant improvement of sensitivity in studies of spin dynamics, notably to resolve spin waves in nanocontacts [55], [56]. An interesting development with STXMs is the nanoXAS beamline at the Swiss Light Source, where the combination of an AFM probing the surface topology, while at the same time using the penetrating x-rays with the STXM allows probing the bulk properties of the specimen [57].

Since both TXM and STXM are photon-in/photon-out microscopy techniques, they can be operated in in principle unlimited external magnetic fields, which enables to follow the evolution of spin textures throughout a complete hysteresis cycle. Similarly, recording magnetic nanostructures and spin patterns across temperatures, e.g. through phase transitions does not pose any fundamental limitations, but has as of today only been realized in a limited range.

One of the requirements so far for transmission soft x-ray microscopies are x-ray transparent substrates. In general those are thin (30-200nm) Si_3N_4 substrates that have been developed and are commonly used for transmission electron microscopies. This prevents e.g. magnetic x-ray microscopy studies of epitaxially grown thin films. Detecting the luminescence that is generated by the focused x-ray spot of a STXM in a substrate, that is otherwise opaque to x-rays, can overcome this limitation [58].

3.2. X-ray photoemission electron microscopy

Whereas TXM and STXM detect the varying x-ray absorption throughout the bulk of the sample, the X-PEEM technique provides magnetic information down to a few nm thin layer below the surface of the sample [59] [60]. As in TXM/STXM a beam of circularly polarized x-rays of a particular photon energy illuminates the sample, however, in X-PEEM the secondary electrons generated in the primary absorption process that penetrate from within the sample to the surface are released by a potential difference applied between the surface and the first electron lens in the electron microscopy column. Therefore, the probing depth in X-PEEM is limited to the escape depth of the electrons, which amounts to a few nm only. The photoelectrons leaving the surface are then propagating through the electron column of the X-PEEM onto a CCD detector. X-PEEM and TXM are therefore both full-field techniques, however, in X-PEEM the electron optics and thus, e.g. the magnification can be easily adjusted, whereas in TXM there is no “adjusting” of magnification without mechanically changing the x-ray optics. The spatial resolution of X-PEEM is limited by aberration in the electron path throughout the system and the latest aberration-corrected X-PEEMs are approaching the 10nm regime as well. Beyond pure increase in spatial resolution, it is even more the high throughput of the latest X-PEEM systems, which can be seen as a major step forward [61].

Time resolved X-PEEM studies is very similar to the analogue studies with TXM (see above). Since X-PEEM work typically in UHV conditions, high frequency or short pulse width electronic pulses cannot be as easily launched to the sample as it is with TXMs, where the sample can be place in ambient conditions. However, the use of laser-based Auston switches allowed for a realization of ultrafast electronic excitations of spin dynamics in confined nanostructures with time resolved X-PEEM, such as vortex gyrations [62]. Recent work in ultrafast spin dynamics has also used X-PEEM to study the magnetization reversal in a ferrimagnet upon ultrafast laser excitation [63],[64].

To retrieve the 3D spin configuration with X-PEEM an interesting Transmission-X-PEEM, also called shadow PEEM approach has been reported recently [53] [65][66][67]. There, the x-rays transmit the object of interest that is mounted above a surface at an oblique angle. The transmitted intensity of the photons reflects the local absorption of the specimen and, when being absorbed in the subsequent substrate, generates photoemission electrons which intensity is proportional to the transmitted intensity of the original photons. Since the footprint of the photon spot is large due to the shallow angle, the image of the electrons from the substrate contains not only the density of the specimen, but allows also to increase the spatial resolution of the transmitted x-rays. Similar to the TXM tomography approach, the specimen is rotated so as to collect a full angular set of projections. From those data, the full 3D magnetic tomography can be retrieved.

To study interfaces X-PEEM has been used in connection with X-ray standing waves, although the depth sensitivity is always limited by the escape depth of the electrons. Using hard or tender x-rays, the probing range can be slightly enlarged.

3.3. Coherent diffraction imaging techniques

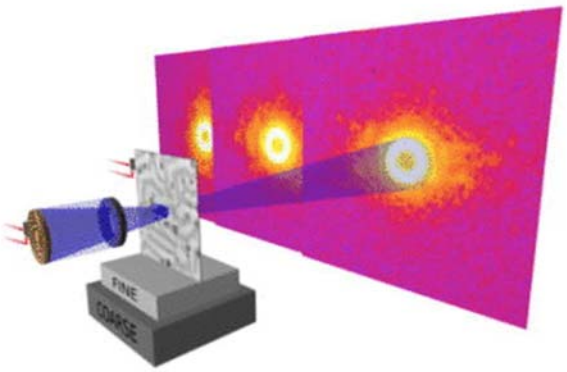

Figure 2: Schematic setup for x-ray ptychography. Reprinted from [31], with the permission of AIP Publishing.

In contrast to the real space imaging techniques described so far, there is a recent push towards imaging techniques working in reciprocal space [68], [69]. They use the diffraction of x-rays, where the local information is encoded both into the amplitude and the phase of the diffracted light. Those techniques rely heavily on the coherence of x-rays, and therefore they will significantly benefit from future developments towards high coherence x-ray light sources in diffraction limited storage rings (DLSR) and XFELs.

3.3.1. Resonant soft X-ray ptychography

The technique of resonant soft x-ray ptychography raster scans with a STXM the sample at a specific wavelength and records for every scan step a full diffraction pattern leading to a high oversampling of the data (Fig. 2). The concept of ptychography is to apply sophisticated algorithms and software packages that allow to retrieve from those vast amount of diffraction patterns both the values of the resonant contribution to the real and the imaginary parts of the refractive image and thus the real space image at high spatial resolution [70], [71]. Standard Fourier Ring Correlation analysis allows to determine the spatial resolution and a 10 nm value for magnetic imaging has been reported [31]. The spatial resolution depends strongly on the scattering contrast of the sample, as well as systematic perturbations to the recorded data. Those include sample positioning errors, drifts, but also unstable illumination, which should drastically improve at the next generation diffraction limited storage rings, where the coherent flux is expected to increase by up to a factor 1,000.
Recently, first approaches to retrieve 3D magnetic tomography information from hard x-ray ptychography experiments have been reported [72], [73].

3.3.2. Holography

Another promising approach of x-ray microscopy with coherent x-rays is Fourier transform X-ray holography (FTH). Similar to optical holography, a real space image is retrieved from the Fourier transform of a hologram originating from the interference of a coherent wave front through the sample with a reference beam that is generated by a narrow and precise reference hole near the object [69], [74], [75] (Fig. 3). FTH has also been shown to work with single shots from an XFEL [32]. However, the spatial resolution of FTH is currently in the 50 nm regime [76], which is still far from the diffraction limit of soft x-rays, mainly because of the challenges of fabricating a sufficiently small and precise reference hole that largely determines the spatial resolution. Further, shrinking the diameter of the reference hole increases the resolution but also limits the available flux and poses significant challenges to the nanopatterning fabrication process itself.

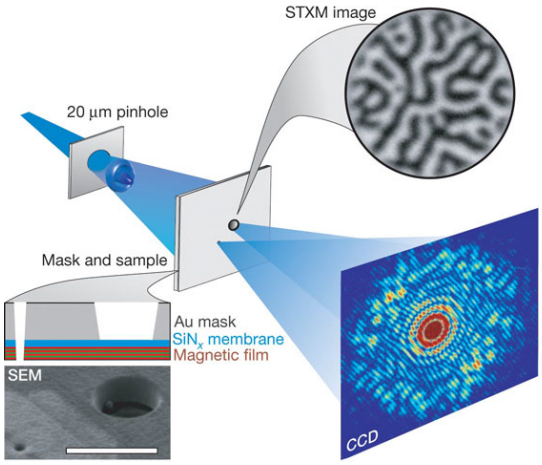

Figure 3: Schematics for an x-ray holography experiment. The lower inset shows the geometry and an electron microscopy image of the mask-sample structure. The top inset is a direct image of the sample recorded with STXM. Reprinted by permission from Macmillan Publishers Ltd: Nature [69], copyright (2004).

The field of view for both ptychography and FTH is rather limited to a few micrometers only, although both techniques can in principle scan larger areas at the expense of recording times. Imaging various orientations of the magnetization or even 3D imaging seems to be rather challenging particularly for FTH.

3.4. Synchrotron X-ray Scanning Tunneling Microscopy

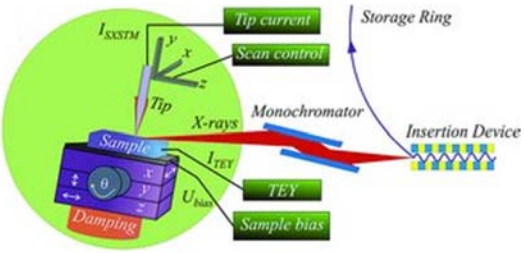

Figure 4: Schematics of Synchrotron X-ray Scanning Tunneling Microscopy. From [77].

Scanning tunneling microscopy (STM), in particular its spin polarized version (SP-STM) has shown to be able to image spin systems with atomic resolution [9]. Combining STMs and SP-STMs with the chemical and magnetic contrast with polarized x-rays from synchrotrons has the exciting perspective to study novel materials in a completely new ways [77]. Fig. 4 shows the schematics of such an experiment at the APS in Argonne IL. The experimental realization of such a systems poses significant challenges, as e.g. the high brillance of x-rays irradiating the sample's surface can impact the tunneling conditions in the tip.

Very recently, a first X-STM study was able to detect the local XMCD spectra at the Fe $L_{2,3}$ edges in a thin Fe film grown on Cu(111) through a tunneling smart tip that served as a photoelectron detector. The study revealed intensity variations in the photo-excited tip current that is indicative of chemical variations within a single magnetic Fe domain [78].

4. Examples from recent research

In the following some specific examples from recent research are provided to demonstrate the current state-of-the-art in magnetic imaging with polarized soft x-rays. The selections are by no means attempting to be comprehensive, but they are selected based on most recent achievements and in view of the most challenging scientific topics for the future, where the most impact can be expected with polarized soft x-rays.

4.1. Towards fundamental magnetic length scales

Fig. 5 shows a MTXM image recorded at the Co L_3 edge at 777eV of a nanogranular 50nm thin $(Co_{83}Cr_{17})_{87}Pt_{13}$ alloy film that was co-sputtered from a CoCr alloy target with Pt chips onto a 40 nm thin Ti buffer layer on a 200nm thick Si_3N_4 substrate to allow for sufficient x-ray transmission through the overall stack. An average grain size of about 35nm of this sample was retrieved from TEM analysis. The inset shows an intensity profile across an area in the image as indicated by the red dotted line. The transition range from a dark to a bright area, i.e. the domain wall between domains with the magnetization pointing up and down, resp. was derived to be at the resolution limit of 15 nm, which is what the FZP optics was expected to deliver. Recording the MTXM images throughout the full hysteresis cycle allowed with this study to determine the local magnetization behavior at the fundamental granular level [79].

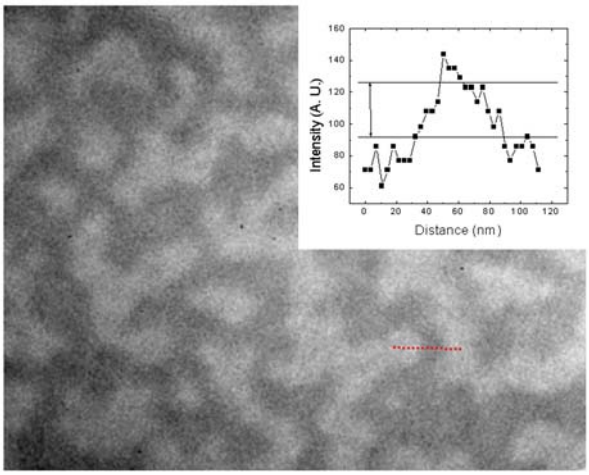

Figure 5: MTXM image of the magnetic domains in a CoCrPt thin nanogranular film. Line intensity profile of the red dotted line in the image is shown in the inset.
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The current state-of-the-art to resolving spin textures on the nanoscale with resonant soft x-ray ptychography is shown in Fig. 5 [31]. An amorphous 50 nm thin SmCo5 film prepared by off-axis pulsed laser deposition onto a similar 200nm thick Si_3N_4 substrate as in the example above was probed with this imaging technique. The ptychography scans consisted of sets of 20x20 diffraction patterns with a 50nm spacing to cover about a 1μm large field of view. At each scan point two diffraction patterns were collected with 10ms and 400ms exposure times to cover regions of higher signal but low momentum transfer and those with lower signal but high momentum transfer, resp. This allowed to extend the

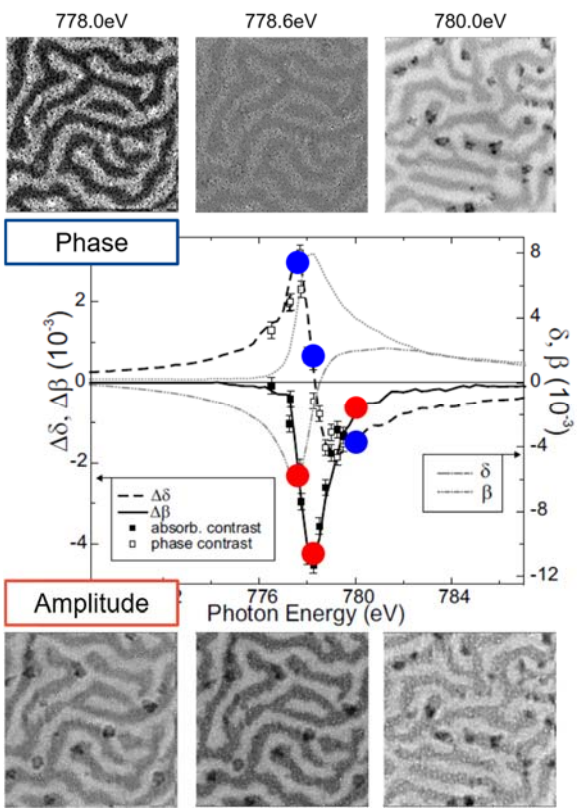

Figure 6: Reconstructed magnetic phase (top) and amplitude images at three photon energies around the resonant Co L_3 edge [31]. The measured contrasts (red and blue dots) match the expected values as shown in the diagram [30].

dynamic range of the measurements and also to improve the spatial resolution. Fig. 6 shows the reconstructed magnetic amplitude and phase contrast images recorded at three photon energies around the resonant Co L_3 edge. The diagram in Fig. 5 displays the real (δ) and imaginary (β) part of the refractive index, which are connected through a Kramers-Kronig transformation. Also shown are the dichroic contributions to δ and β, which correspond to the magnetic amplitude and phase contrast. The magnetic amplitude contrast follows the well-known XMCD profile with the maximum contrast close to the largest x-ray absorption (red dots), whereas the magnetic phase contrast (blue dots) has the largest contrast before and after the resonant edges with a sign reversal going through the resonance [30]. At resonance the phase contrast vanishes. Magnetic phase contrast has so far not yet been fully exploited, although the large magnetic contrast at lower absorption could be advantageous at high intensity XFELs to reduce sample damage. Of even larger importance could be the magnetic phase contrast at high coherence x-ray sources which allow to develop x-ray interferometric measurement techniques. Those will offer much higher sensitivity and accuracy to characterize e.g. highly diluted magnetic systems. The spatial resolution obtained in the images in Fig. 6 was determined from Fourier Ring Correlation analysis yielding a value of about 10 nm, which is not yet sufficient to determine e.g. the internal spin texture of the domain walls in those thin films.

A recent magnetic x-ray transmission spectromicroscopy study performed with a multilayered (Co 0.3 nm/Pt 0.5 nm) x 30 heterostructure across the Co L_3 and L_2 edges has shown a reduced L_3/L_2 XMCD signal, which could be explained by a spin texture inside the domain wall, that would be much more complex than the commonly assume Bloch wall [80]. To experimentally verify this hypothesis, a full 3D tomographic characterization of the internal spin texture is needed.

4.2. Towards magnetic x-ray tomography

Understanding the fundamental building blocks of magnetic materials at the nanoscale is one thrust in magnetism research, however, it was recently recognized that mesoscale phenomena, i.e. properties, behavior and functionality at larger length scales have also be taken into account. Investigations that reach across length scales are essential to understand e.g. complexity and hierarchies in novel materials.

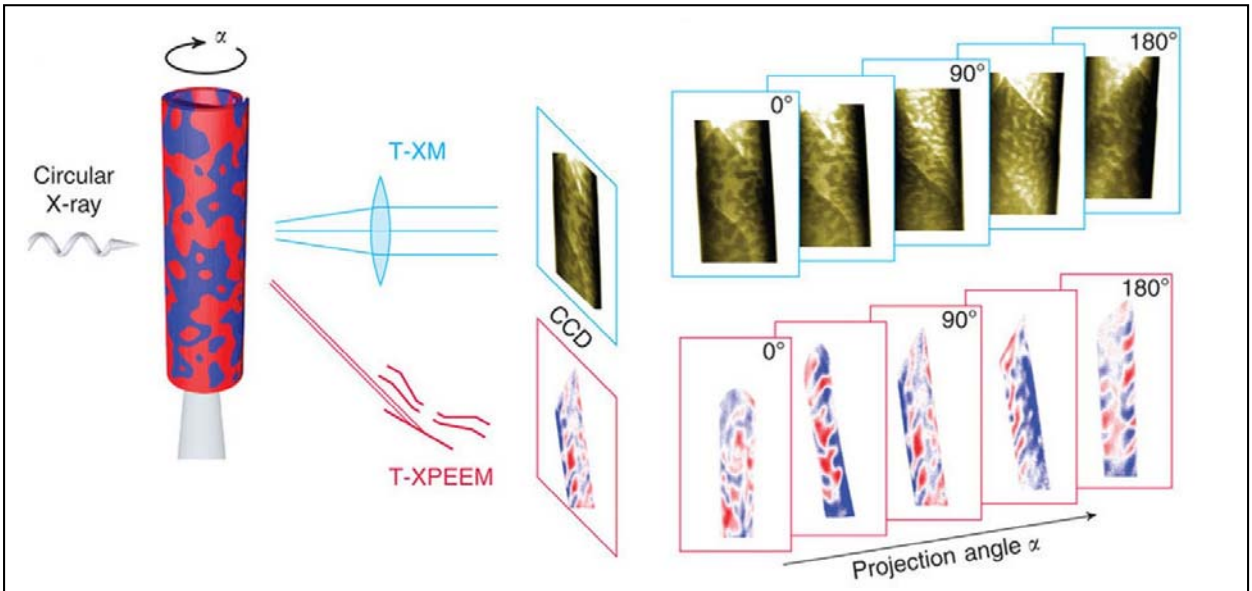

Figure 7: Schematic showing the concept of magnetic x-ray tomography recorded with MTXM (top) and X-PEEM (bottom). A set of images at various rotation angles is shown in the right half of the figure. From [53].

Those effects can occur e.g. in three dimensional magnetic structures that harness in a complete way the full vectorial character of the spins [81]. The ultimate goal will be to achieve multidimensional characterization with nanoscale (fundamental) spatial resolution that will provide unique insight and key information into complex spin configurations. An extension of x-ray spectro-microscopies seems to provide again a unique gateway to achieve those goals, combining penetration power with spatio-temporal resolution, and again elemental sensitivity. Once such capabilities become available, questions, such as the switching of a magnetic nanowires, the spin configuration in core-shell nanoparticles, or the spin textures in a propagating domain wall along a nanowire can be addressed and answered. Full-field x-ray microscopies, specifically MTXM and X-PEEM have recently shown first successful results employing a tomographic concept, which is schematically depicted in Fig. 7 [53]. The system studied was a tubular thin magnetic Co/Pd film architecture, which was fabricated by strain engineering. The 3D arrangement of the spin textures was retrieved by recording an angular set of 2D projections that in case of MTXM were directly imaged by the X-ray optics, whereas in the case of X-PEEM the x-rays transmitted the system and generated photoelectrons from the substrate. These results allowed to obtain quantitative information about domain patterns and magnetic coupling phenomena between windings of azimuthally and radially magnetized tubular objects.

4.3. Looking at buried magnetic interfaces

One of the most challenging topics in characterization magnetic materials is to understand the role that buried interfaces play for the properties, dynamics, and overall functionality of magnetic systems. Getting direct and non-destructive access to those interfaces is therefore of paramount scientific and technological interest. One approach is based on the use of x-ray standing waves to probe deep into the materials and thus achieve depth sensitivity. In combination with a laterally resolving microscopy, such

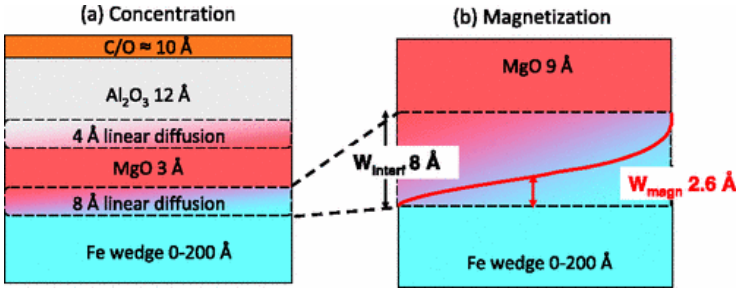

Figure 8: Results from x-ray standing wave experiments to determine the concentration (a) and magnetization profiles (b) across an Fe/MgO interface. Reprinted figure with permission from [39].

as X-PEEM a full 3D information, in particular about the buried interfaces can be obtained. In a recent study this was applied to an array of circular magnetic Co nanodots that were deposited on a multilayer substrate with the composition of (23.6 Å-Si/15.8 Å-Mo)x40, which served as the generator of an x-ray standing wave. The latter was moved vertically through the sample by tuning the photon wavelength around the Bragg condition. Thus depth resolved information at a resolution determined by the accuracy of the incoming photon energy can be retrieved. The emitted photoelectrons were imaged with X-PEEM at better than 100nm lateral spatial resolution and an analysis of their energy dependence and comparison to x-ray optical calculations allowed to quantitatively derive the depth-resolved film structure of the sample and various coatings using the element-specificity of x-rays [37]. Fig. 8 shows results from an X-ray standing wave experiment of an Fe/MgO tunnel junction, where the magnetization profile across the Fe/MgO interface could be retrieved with sub-A resolution [39].

A particular buried structure are topological defects in bulk systems that impact the behavior and functionality of materials. With the penetration power of soft x-rays of about 100-200nm and anticipated spatial resolutions below 10nm, imaging those defects at varying angles, similar to stereo-microscopy approaches in optical systems, allows to characterize them. A recent investigation with MTXM on a 55–120 nm thick ferromagnetic NdCo5 layers retrieved complex topological defects by using the angular dependence of magnetic contrast in a series of high resolution MTXM images. These findings and instrument capabilities open a path for a systematic characterization of deeply buried magnetic topological defects, nanostructures and devices [82].

4.4. Imaging spin dynamics

Beyond the nanoscale spatial information described above, which provides deep insight into the static and ground state properties of spin textures, the functionality of magnetic materials, i.e. the dynamics behavior of spin textures is of paramount importance. The inherent time structure of x-ray pulses generated at either electron storage rings or XFELs spanning from the fsec regime at XFELs to the ps and ns regime provide unique information for studies of spin dynamics. At storage rings the number of photons per electron bunch is far too low to allow for single shot imaging, and therefore commonly pump-probe experiments are being performed. One of the challenges is that in order to detect small changes in the magnetization on short time scales and nanometer spatial scales requires a stable and precise locking of the excitation pump pulses with the probing x-ray pulses. A major step forward has recently been implemented at the STXM beamline at the Stanford Synchrotron Radiation Lightsource

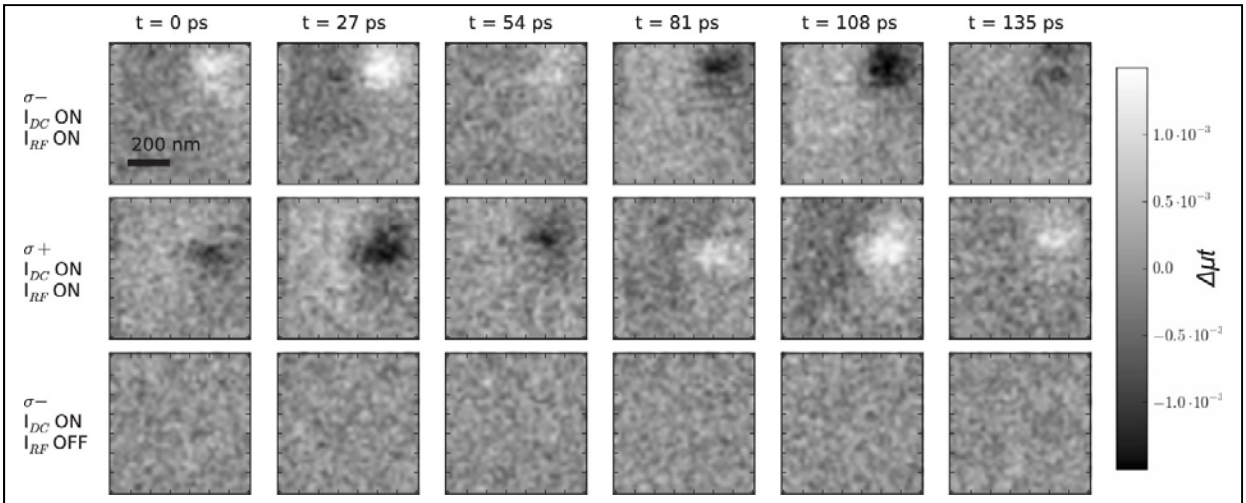

Figure 9: Time resolved magnetic STXM images of the spin dynamics in a spin torque oscillator. The first two rows show the results at various delay times between pump and probe, i.e. at various phases of the magnetization precession. The third row are data recorded with the RF current switched off.

(SSRL) in Stanford CA by a novel microwave synchronization scheme [56] to study specifically high frequency magnetization dynamics occurring e.g. as spin waves generated by a spin torque oscillator [83]. Direct images of the spin dynamics in the 5-10GHz range can be acquired with a spatial resolution in the few tens of nanometer regime using XMCD as magnetic contrast. A quasi-stroboscopic detection scheme allows to compensate e.g. drifts that generally limit the sensitivity. With a jitter of about 500fs and a drift of 2ps over 1h, which is small compared to the temporal width of the x-ray pulse (50ps), extended measurement times are possible that enable to image the dynamics in the spin waves emitted by a nanocontact spin torque oscillator at 6.3GHz frequency and the emergence of a localized soliton with a nodal line, that is, with p-like symmetry[84], as well as the FRM amplitude of only ~0.1° at 9.1GHz in a micronsized Co strip[56]. Fig. 9 shows the time-resolved XMCD images of the magnetization dynamics in the spin torque oscillator that is triggered by a high-density, spin-polarized current injected via a nanometer size contact into an extended ferromagnetic film.

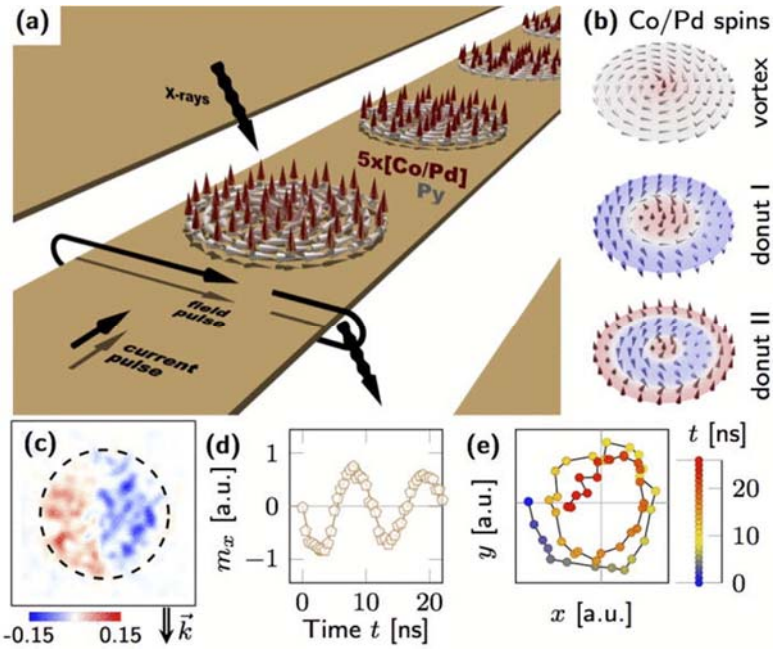

Figure 10: a) Schematic diagram of the experimental setup to measure the time-resolved dynamics with MTXM. b) Simulated spin configuration in the Co/Pd multilayer stacks as a function of the interlayer coupling. (c) Visualization of in-plane magnetization component of the Py subsystem utilizing XMCD. (d) Extracted oscillations of the magnetization component that are correlated to the vortex core displacement (e).
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Fig. 10 shows the schematics and experimental results of a time resolved MTXM study on the magnetization dynamics of imprinted non-collinear spin textures [85]. There, the magnetic vortex state of a soft permalloy micron-sized disk was imprinted into a magnetically hard Co/Pd nanopatterned heterostructure with a pronounced out-of-plane magnetization. Previous static MTXM studies have shown that the interlayer exchange coupling between the soft and the hard magnetic disc structures allows to tailor the magnetic state in the Co/Pd structure, e.g. from the vortex (strong coupling) to a donut-type (weaker coupling) spin texture. Since the vortex and the donut-type texture have different topological winding numbers that are characteristic e.g. in skyrmion textures, this approach allowed to generate magnetic skyrmions at room temperature through a proper design of the interlayer coupling [86]. Studying the dynamics in a pump-probe stroboscopic setup with MTXM revealed the appearance of two distinct gyration frequencies that occurred at the early and the later stages in the dynamics, resp. Furthermore the high spatial resolution images indicated a widening of the size of the vortex core compared to the original magnetic vortex that was confirmed by micromagnetic simulations using the Thiele equation [85].

5. Conclusion and Outlook

Magnetic imaging with polarized soft x-rays has become a powerful and indispensable tool to acquire a deeper understanding of static properties, dynamic behavior and novel functionalities of spin textures in novel magnetic materials. The magnetic x-ray spectro-microscopies combine in a unique way spatial and temporal resolution that can reach into the fundamental limits of magnetism but can also bridge across spatio-temporal scales. Single shot x-ray imaging has started recently harnessing the high peak intensities per pulse at XFEL sources. A diffraction limited spatial resolution that for x-rays is genuinely in the few nanometer regime, has not yet been demonstrated with magnetic materials, however, novel x-

ray microscopies harnessing specifically the features of next generation x-ray sources should be able to reach that goal. Characterizing buried interfaces and particularly their dynamics, e.g. observing the effect of a spin current running through an interface on the spin textures at the interface is probably one of the experimentally most demanding, but scientifically most rewarding topics. Pushing magnetic x-ray imaging into the 3rd dimension is a very promising new direction that could have significant impact to synthesizing novel materials and discovering new applications. The key feature of any magnetic x-ray spectromicroscopy is that an analysis of the element-specific dichroic contrast is a quantitative measure of magnetic ground state properties of individual components, notably the spin and orbital magnetic moments in a multicomponent system. In the search for novel magnetic materials, a spatially and time resolved characterization, tailoring and understanding of those fundamental quantities will be of paramount importance.

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